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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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<p>(21) International Application Number: PCT/US98/00325 (22) International Filing Date: 15 January 1998 (15.01.98) (30) Priority Data: 60/035,509 15 January 1997 (15.01.97) US (71) Applicant: MICROBIOMED CORPORATION [-/US]; 2454 Walnut Ridge Street, Dallas, TX 75229 (US). (72) Inventors: UTECHT, Ronald, E.; P.O. Box 19, Volga, SD 57071 (US). LEWIS, David, E.; 816 3rd Avenue, Eau Claire, WI 54703 (US). (74) Agent: CHWANG, T., Ling; Hitt Chwang &amp; Gaines, P.C., 225 University Plaza, 275 West Campbell Road, Richardson, TX 75080 (US).</p>		<p>(81) Designated States: AL, AM, AT, AU, AZ, BB, BG, BR, BY, CA, CH, CN, CZ, DE, DK, EE, ES, FI, GB, GE, HU, IL, IS, JP, KE, KG, KP, KR, KZ, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, TJ, TM, TR, TT, UA, UG, UZ, VN, European patent (AT, BE, CH, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent (BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, NE, SN, TD, TG).</p> <p>Published With international search report. Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</p>
<p>(54) Title: NAPHTHALIMIDE DYES AND THEIR USE AS FLUID VISUALIZING AGENTS</p> <div data-bbox="232 1140 1211 1449" data-label="Chemical-Block"> </div> <p>(57) Abstract</p> <p>The present invention provides an amino-1,8-naphthalimide compound having substituent groups at the imide position and a position distal to the imide position. The substituent groups are linked to a hydrocarbon directly linked at the imide position and the distal position. The substituent groups render the amino-1,8-naphthalimide compound sufficiently soluble in a fluid such that visualization of the fluid by color or fluorescence is allowed. In one advantageous embodiment, the amino-1,8-naphthalimide is obtained from the mixture of 4-chloro-1,8-naphthalic anhydride with monoamine polyether.</p>		

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## NAPHTHALIMIDE DYES AND THEIR USE AS FLUID VISUALIZING AGENTS

## BACKGROUND OF THE INVENTION

5           Various processes for fluid detection in  
hermetic and semi-hermetic systems are well  
known. In such systems, it is highly desirable  
to determine when a leak occurs in the system to  
prevent further loss of the operating fluid and  
10   its ultimate failure. Difficulty arises,  
however, in detecting the various fluids that  
might be employed in these sealed or semi-sealed  
systems. For example, the fluid may be a  
colorless gas that is impossible to visually  
15   detect, such as a gas used in a refrigeration  
system. Alternatively, the fluid may be one that  
combines with other fluids to the extent that its  
identity is difficult to visually determine, such  
as a fluid that leaks from a piece of machinery  
20   and combines with other residual fluids that have  
been deposited on the machinery. To enable  
service technicians to more easily detect the  
leakage of these fluids, various dyes have been  
developed that can be added to the fluid. The  
25   dyes typically will fluoresce when excited by the  
appropriate wavelength of light. Over the past  
several years, environmental concerns have arisen  
regarding the usage of certain hermetic fluids,  
such as the fluor-hydrocarbon gases typically  
30   used in earlier refrigeration systems.

In response to these environmental concerns, hermetic and semi-hermetic fluid systems that are environmentally safe have been developed and continue to be developed to replace the fluor-  
5 hydrocarbon compounds used in many of these systems. These new alternative fluid systems typically require the use of new kinds of lubricants such as synthetic polyalkylene glycols (PAG) and polyolesters (POE) and have rendered  
10 conventional leak detection chemicals employing these new lubricants largely ineffective. In response thereto a new generation of fluorescent dyes have been developed that are more compatible with these new lubricants. However, even these  
15 new leak detection dyes suffer from certain disadvantages, as explained below.

It is known that fluorescent dyes, including certain non-amino naphthalimides, have application to the above-discussed leak detection  
20 in lubrication and refrigerant systems and other hermetic and semi-hermetic systems. Basically, the dye is added to the lubricant or refrigerant and if leaks occur in the system that contains the lubricant or refrigerant, they can be  
25 visually identified by exciting the leaked dye/lubricant/refrigerant mixture with a suitable light source, which is typically an ultra violet (UV) light source. It is important to note that, particularly in refrigeration systems, the  
30 lubricant mixes with the refrigerant in the course of operation. Accordingly, a leak detection dye added to the lubricant will nonetheless work to detect leaks in the refrigerant/and visa versa.

In many such applications, visualization of fluids, fluid leakage, and the mode of fluid leakage from a system, is difficult due to the clarity of the fluid and the surrounding fluid puddle. Various colored and fluorescent materials have, therefore, been made as additives which allow the visualization of fluids. Currently available compounds, however, suffer from a lack of suitable solubility in some fluids such as polyethyleneglycol based fluids.

This particular disadvantage has been overcome, but only at the expense of adding a relatively large amount of certain naphthalimide dyes first to a small portion of the system's fluid or a suitable solvent and then adding the pre-mixed mixture to the fluid. Unfortunately, however, in these applications, a relatively large amount of naphthalimide is required to generate a visible fluorescent signal. Moreover, most conventional leak detection dyes require the use of ultra violet (UV) excitation, which means that expensive, bulky UV light sources must be used by field personnel to perform leak detection diagnostics. Health and safety risks attendant with the use of UV light are also a disadvantage.

Non-azo, hydrophobic naphthalimide dyes have also been developed but have specific use in the biochemical area. These dyes may be combined with an activating agent to generate an activated species. The activated species can initiate chemical changes in a lipid bilayer membrane of a virus or another target cell. These changes can eradicate viruses and other target cells and cause structural changes in the lipid and any

associated proteins. While these non-azo, hydrophobic naphthalimide dyes have excellent application in the field of biochemistry they are poorly suited for application in the detection of fluid leakage in a hermetic or semi-hermetic system. A primary reason for this is that they are typically hydrophobic compounds that do not have a high degree of solubility in polar solvents or fluids, such as those typically used in hermetic or semi-hermetic systems. As such, their application in such systems is undesirable because solubility of the detection dye in the fluid is an important factor in providing an efficient leakage detection compound.

Accordingly, what is needed in the prior art is a compound that is sufficiently soluble in a fluid such that substantially less amounts of the compound are required and that visualization of the fluid by color or fluorescence is allowed without the use of UV light sources.

## SUMMARY OF THE INVENTION

To overcome deficiencies in the prior art, the present invention provides, in one embodiment, a 3 or 4-amino-1,8-naphthalimide compound having polyether substituent groups at the 3 or 4 position and the imide position. Preferably, the polyether groups are linked to a hydrocarbon directly linked at the 3 or 4-amino position and the imide position. However, it will, of course, be appreciated by those skilled in the art that compounds linked at the 3 or 4 position also include those compounds where the substituent groups are linked at the 5 or 6 positions.

The polyether substituent groups render the 3 or 4-amino-1,8-naphthalimide compound sufficiently soluble in a polar fluid such that visualization of the fluid by color or fluorescence is allowed. In one advantageous embodiment, the 3 or 4-amino-1,8-naphthalimide is obtained from the mixture of 4-chloro-1,8-naphthalic anhydride with monoamine polyether.

In another embodiment, the present invention provides an amino-1,8-naphthalimide compound that is soluble in a fluid and that has substituent groups at an imide position and a position distal to said imide position. The substituent groups are linked to a hydrocarbon directly linked at the imide position and the distal position. Additionally, the substituent groups have chemical properties that render the amino-1,8-naphthalimide compound sufficiently soluble in

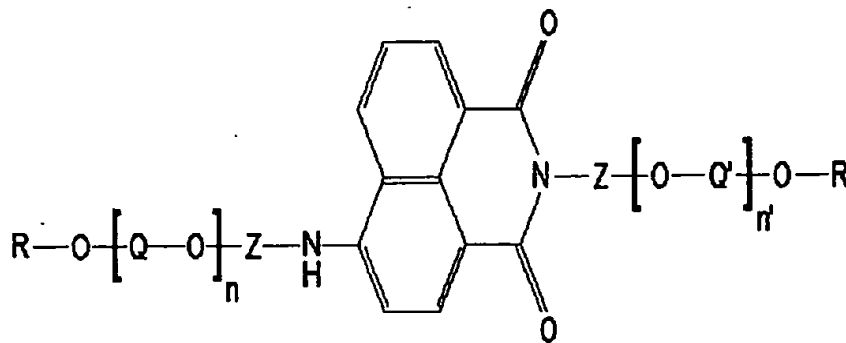


the fluid such that visualization of the fluid by color or fluorescence is allowed.

The present invention, therefore provides, in a broad scope, a colored material (chromophore) or a fluorescent material (fluorophore) compound having substituents thereon that renders the material sufficiently soluble in a fluid so that visualization of the fluid by color or fluorescence is allowed. The substituents used are preferably chemically and physically identical or sufficiently similar in properties to the fluid that the chemical and physical forces of solvation lead to sufficient solubilization to allow detection of the dissolved colored or fluorescent material by visual or by light-detector means.

In another embodiment of the present invention, the fluid is a polyalkylene glycol lubricant, and in such embodiments, the polyalkylene glycol is preferably polyethyleneglycol. In this particular embodiment, the 4-chloro-1,8-naphthalic anhydride may comprise about 2% by weight of the 4-amino-1,8-naphthalimide compound and the monoamine may comprise about 97% by weight of the 4-amino-1,8-naphthalimide compound. In another aspect of this particular embodiment, the mixture is heated to approximately 150°C for 48 to 72 hours.

In another embodiment, the amino-1,8-naphthalimide compound has the following general chemical formula:



Where:

R or R' may be hydrogen, hydrocarbon, or halogenated hydrocarbon and where R may or may not equal R' ;

5        Q or Q' may be hydrocarbon or halogenated hydrocarbon and where Q<sub>n</sub> may or may not be equal to any other Q or Q' ; and

      Z or Z' may be hydrocarbon or halogenated hydrocarbon and where Z may or may not equal Z' .

10       In each occurrence in the above-referenced structure, n may range from 0 to 200. In a more specific embodiment, however, n may range from 9 to 10.

      In another aspect of this particular embodiment, the hydrocarbon is a halogenated hydrocarbon.

15       Another aspect of the present invention provides a 3 or 4-amino-1,8-naphthalimide compound that when mixed with the fluid has a  
20       excitation wavelength spectra of centered of approximately equal to or greater than 400 nm.

      In yet another aspect of the present invention, the substituents groups are polyether groups that have the following general formula:

25        $\text{CH}_3\text{--}[\text{O--CH}_2\text{--CH}(\text{CH}_3)]_n\text{--}$

      where n may range from 5 to 15.

In another aspect of the present invention, there is generally provided a method of detecting the fluorescent emission spectrum of a detection mixture. In a preferred embodiment, the method  
5 comprises the steps of mixing a 3 or 4-amino-1,8-naphthalimide with a polyether substituent to form a mixture having polyether substituent groups at a 3 or 4-position and at an imide position. The polyether substituent groups are  
10 linked to a hydrocarbon directly linked at the 3 or 4-position and the imide position. The polyether substituent groups render the 3 or 4-amino-1,8-naphthalimide compound sufficiently soluble in a fluid such that visualization of the  
15 fluid by color or fluorescence is allowed. The method further comprises the steps of mixing the mixture with the fluid to form the detection mixture, irradiating the detection mixture with a light sufficient to cause the detection mixture  
20 to emit a detectable excited wavelength emission spectrum, and detecting the emission spectrum or fluorescence of the detection mixture.

The foregoing has outlined, rather broadly, preferred and alternative features of the present  
25 invention so that those who are skilled in the art may better understand the detailed description of the invention that follows. Additional features of the invention that form the subject of the claims of the invention are  
30 described below. Those who are skilled in the art should appreciate that they can readily use the disclosed conception and specific embodiment as a basis for designing or modifying other structures for carrying out the same purposes of

the present invention. Those who are skilled in the art should also realize that such equivalent constructions do not depart from the spirit and scope of the invention in its broadest form.

## BRIEF DESCRIPTION OF THE DRAWINGS

For a more complete understanding of the present invention, reference is now made to the following descriptions taken in conjunction with the accompanying drawings, in which:

FIGURE 1 illustrates a specific chemical structure of a preferred embodiment of the compound covered by the present invention;

FIGURE 2 illustrates a chemical structure of a conventional naphthalimide core;

FIGURE 3 illustrates a chemical structure of a polyalkylene glycol;

FIGURE 4 illustrates a fluorescence spectra of a CI Yellow 143 dye and a MBM Yellow JM6 dye in a polyalkylene glycol lubricant at a concentration of  $1\mu\text{M}$ .

FIGURE 5 illustrates the absorbance spectra of a CI Yellow 143 dye and the MBM Yellow JM6 in a polyalkylene glycol lubricant at a concentration of  $10\mu\text{M}$ ; and

FIGURE 6 illustrate a general chemical of a preferred compound covered by the present invention.

## DETAILED DESCRIPTION

As previously discussed, the present invention provides, in one embodiment, a 3 or 4-amino-1,8-naphthalimide compound having polyether  
5 substituent groups at the 3 or 4 position and at the imide position that render the compound readily soluble in a polar fluid. The preferred substituents used to form the compound are chemically and physically identical or  
10 sufficiently similar in properties to the target fluid (i.e., the fluid used in the intended hermetic or semi-hermetic system) that the chemical and physical forces of solvation lead to inherent solubilization to allow detection of the  
15 dissolved colored or fluorescent material by visual or by light-detector means. By way of example only, such fluids, may include polyalkylene glycols (PAG), polyolesters (POE), polyethylene glycols (PEG), mineral oils, engine  
20 oils, gear and transmission oils and greases, hydraulic fluids, hydro-chlorofluorocarbons, hydrofluorocarbons, or chlorofluorocarbons and refrigerants such as R-134a.

In another embodiment, the present invention  
25 provides an amino-1,8-naphthalimide compound that is soluble in a fluid and that has substituent groups at an imide position and a position distal to said imide position. The substituent groups are linked to a hydrocarbon directly linked at  
30 the imide position and the distal position. Additionally, the substituent groups have chemical properties that render the amino-1,8-

naphthalimide compound sufficiently soluble in the fluid such that visualization of the fluid by color or fluorescence is allowed.

5 In a more preferred embodiment, however, the polyether groups are linked to a hydrocarbon directly linked at the 3 or 4-amino position and the imide position. In general, the substituent groups render the 3 or 4-amino-1,8-naphthalimide compound inherently soluble in a polar fluid such  
10 that visualization of the fluid by color or fluorescence is allowed. This is in contrast to many of the conventional dyes, which typically forms suspensions and not true solutions. Thus, the present invention provides a compound that is  
15 hydrophillic and easily soluble in fluids, such as those typically used in hermetic and semi-hermetic systems.

Because of the compound's enhanced solubility properties, it is not necessary to  
20 pre-mix the compound of the present invention with the target fluid or a solvent such as alcohol as required by many conventional compounds. Moreover, as explained below, the unique chemical compound covered by the present  
25 invention, when combined with the target fluid, readily provides a mixture that will fluoresce without the need of excitation by UV light as required by detection dyes disclosed in the prior art. In one particular advantageous embodiment,  
30 the 3 or 4-amino-1,8-naphthalimide may be obtained by mixing 4-chloro-1,8-naphthalic anhydride with monoamine polyether in a manner discussed below.

The present invention, therefore provides in a broad scope, a colored material (chromophore) or a fluorescent material (fluorophore) compound having substituents thereon that render the material sufficiently soluble in a fluid so that visualization of the fluid by color or fluorescence is allowed.

The present invention, in a preferred embodiment, may be formed from the synthesis of the 4-chloro-1,8-naphthalic anhydride with a polyether. The resulting compound is generally illustrated in FIGURE 6 and a more specific embodiment is illustrated in FIGURE 1, which is the naphthalimide-based, yellowish colored, fluorescent molecule MBM Yellow JM-6. This synthesis yields a compound that has enhanced inherent solubility and requires substantially smaller amounts than conventional dyes.

In contrast, the conventional colored fluorescent naphthalimide core with molecular structure shown in FIGURE 2 has only limited solubility in polyethyleneglycol fluids. Additionally, molecular interactive forces between hydrophobic naphthalimides like Figure 2 in hydrophylic polarized solvents cause molecular association of the dye molecules at high dye concentrations. This self association alters the electronic excited states and decreases fluorescence, intensity and efficiency. Therefore, a substantial amount of the compound is typically pre-mixed with the target system's fluid to obtain the proper degree of visualization upon excitation by a UV light source.



With the molecular structure shown in FIGURE 3, the forces of solvation between the large, bulky naphthalimide core of FIGURE 2 and the linear polyether structure of the polyethyleneglycol fluid molecule are too small to attain sufficient concentration for visualization of the naphthalimide color or fluorescence. Synthesis of the substituted naphthalimide shown in FIGURES 1 and 6 incorporates the preferred polyether structure of the fluid, which results in a molecule highly soluble in the polyethyleneglycol fluids.

In tests conducted on one embodiment, incorporation of the polyether substituents did not appreciably alter any of the molecular optical absorption coefficients, the optical absorption spectrum or the fluorescence excitation spectrum of the naphthalimide core as shown in FIGURES 4 and 5.

A nominal 1 micromolar concentration solution of MBM Yellow JM-6 (mol. wt. approx. 1300) in polyalkylene glycol (PAG) lubricant was prepared by direct dilution of JM-6 into the PAG lubricant. Fluorescence emission and excitation spectra were obtained with use of a spectraflorometer. A single excitation maximum was observed at approximately 430 nm and two emission maxima at 497 nm (largest amplitude ( $R_1$ )) and at 525 nm (smaller amplitude ( $R_2$ ,  $R_2/R_1 = 1.15$  approximately)). A solution of the solid (dry) naphthalimide, CI yellow 143, in the PAG lubricant exhibited excitation and emission maxima at nominally the same wavelengths, and within approximately 5% of the same amplitude as

the JM-6 dye. This experiment verified the photophysical integrity of the substituted naphthalimide core in the JM-6 dye.

One family of structural generalizations of the compound MBM Yellow JM-6 (FIGURE 1) is the preferred series of compounds shown generally illustrated in FIGURE 6. However, those who are skilled in the art can appreciate that other chemically equivalent structures of this compound are also within the broad scope of the present invention. For example, the polyether substituent may also be positioned at the 3,5 or 6 positions on the naphthalimide structure, as well as on the imide position.

Solubility of the naphthalimide core in various selected fluids may be adjusted by the selection of the groups R, R', Q, Q', Z, Z', and the numbers n and n' where R may be Hydrogen, hydrocarbon, or halogenated hydrocarbon R may or may not equal R', may be hydrocarbon or halogenated hydrocarbon and Q<sub>n</sub> may or may not be equal other Q or Q' and Z may be hydrocarbon or halogenated hydrocarbon Z may or may not equal Z'.

#### Synthesis of MBM Yellow JM-6

A preferred embodiment of the compound covered by the present invention (MBM Yellow JM-6) was synthesized according to the following procedure:

0.50 grams of 4-chloro-1,8-naphthalic anhydride was added to 20 grams of a monoamine polyether, specifically Jeffamine M-600. The system was then degassed to remove all oxygen and placed under a partial pressure of nitrogen to

prevent oxygen from entering the system. The reaction mixture was heated to approximately 150° for 48 hours after which the mixture was cooled to room temperature under a nitrogen atmosphere.

5 The excess monoamine polyether was removed by extraction with a minimum amount of water.

The presence of the molecular compound MBM Yellow JM-6 of FIGURE 1 at a concentration of 1 $\mu$ M in polyalkylene glycol lubricant enabled

10 visualization by the human eye in a lighted room of a thin film of the fluid dye mixture pressed by fingers between two glass microscope slides upon illumination with the output from a 150 W xenon lamp using a 420 nm interference filter

15 having a maximum transmittance of 20% with the visualization away from the direction of reflected and incident exciting beams.

Incorporation of a colored or fluorescent material with sufficiently enhanced solubility in

20 the fluid of interest by use of the invention will allow visualization or detection of the fluid. For example, a hand-held light emitting a spectrum within the excitation spectrum of the colored or fluorescent material will allow an

25 operator to inspect mechanical system for evidence of lubricant or hydraulic fluid leakage. Alternatively, an electrooptical detector such as a photodiode with suitable filter can be used for continuous surveillance for the presence of a

30 colored or fluorescing fluid.

Solutions of MBM Yellow JM-6 in polyalkylene glycol (PAG) lubricant (Type GPH-6, Interdynamics, Brooklyn, NY) of nominal 1,280 micromolar, 128 micromolar, and 64 micromolar

concentrations were prepared as in Experiment 1. These concentrations correspond to dilutions of 1 to 600, 1 to 6,000 and 1 to 12,000 respectively. Thin films of each of the solutions were smeared separately onto glass microscope slides and an aluminum metal sheet. Each of the films were readily visualized as bright yellow-green, a) in the presence of normal room lighting and b) in normal room lighting and in darkness using filtered blue light obtained with a nominal 80% transmissive 400 - 465 nm interference filter, fitted on a 4-"D" cell sized "Mag-Lite", equipped with a "Mag-Num Star" krypton bulb. This experiment demonstrated the utility of the JM-6 dye under light excitation in the visualization of dye-doped PAG lubricant under conditions mimicking PAG lubricant system leaks. This experiment also demonstrated the feasibility of using simple, economical, and safe light sources for leak detector dye excitation.

The results of the above-example of the present invention demonstrate the utility of the invention. The use of one or more chemical substituents of chemical and physical properties sufficiently similar to the solvating fluid may be linked to a amino naphthalimide dye to attain sufficient solubility of the dye in the fluid to allow visualization or detection of the fluid.

Theoretical organic synthesis and physical organic chemistry considerations show that the polyether substituent structures of the molecule of FIGURE 1 can be extended to include the more general naphthalimide compound of FIGURE 6.

Similar theoretical considerations show that the concept of the invention can be extended to other colored materials and fluorescent materials and fluid materials.

5        From the foregoing, it is readily apparent that the present invention provides an amino-1,8-naphthalimide compound having substituent groups at the imide position and a position distal to the imide position. The substituent groups are  
10        linked to a hydrocarbon directly linked at the imide position and the distal position. The substituent groups render the amino-1,8-naphthalimide compound sufficiently soluble in a fluid such that visualization of the fluid by  
15        color or fluorescence is allowed. In one advantageous embodiment, the amino-1,8-naphthalimide is obtained from the mixture of 4-chloro-1,8-naphthalic anhydride with monoamine polyether.

20        Although the present invention has been described in detail, those who are skilled in the art should understand that they can make various changes, substitutions and alterations herein without departing from the spirit and scope of  
25        the invention in its broadest form.

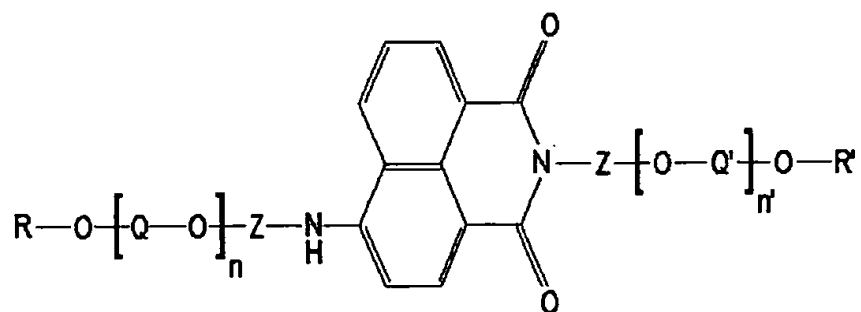
## WHAT IS CLAIMED IS:

1. A 3 or 4-amino-1,8-naphthalimide  
2 compound having polyether substituent groups at  
3 a 3 or 4 position and an imide position, said  
4 substituent groups linked to a hydrocarbon  
5 directly linked at said 3 or 4 position and said  
6 imide position, said substituent groups rendering  
7 said 3 or 4-amino-1,8-naphthalimide compound  
8 sufficiently soluble in a fluid such that  
9 visualization of the fluid by color or  
10 fluorescence is allowed.

2. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 1 wherein said fluid  
3 is a polyalkylene glycol lubricant.

3. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 2 wherein said  
3 polyalkylene glycol is polyethyleneglycol.

4. The 3 or 4-amino-1,8-naphthalimide compound  
 as recited in Claim 1 wherein said 3 or 4-amino-  
 1,8-naphthalimide is a 4-amino-1,8-naphthalimide  
 compound and has the following general chemical  
 structure:



where:  
 R or R' may be hydrogen, hydrocarbon, or  
 halogenated hydrocarbon and where R may or may  
 not equal R';  
 Q or Q' may be hydrocarbon or halogenated  
 hydrocarbon and where Q<sub>n</sub> may or may not be equal  
 any other Q or Q'; and  
 Z or Z' may be hydrocarbon or halogenated  
 hydrocarbon and where Z may or may not equal Z'.

5. The 3 or 4-amino-1,8-naphthalimide  
 compound as recited in Claim 4 wherein n ranges  
 from 0 to 200.

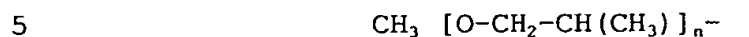
6. The 3 or 4-amino-1,8-naphthalimide  
 compound as recited in Claim 5 wherein n ranges  
 from 9 to 10.

7. The 3 or 4-amino-1,8-naphthalimide  
 compound as recited in Claim 1 wherein said 3 or  
 4-amino-1,8-naphthalimide is hydrophillic.

8. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 1 wherein said  
3 hydrocarbon is a halogenated hydrocarbon.

9. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 1 wherein said 3 or  
3 4-amino-1,8-naphthalimide compound when mixed  
4 with said fluid has an excitation wavelength  
5 spectra equal to or greater than 400 nm.

10. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 1 wherein said  
3 substituent groups are polyether groups that have  
4 the following general formula:



6 where n may range from 5 to 15.

11. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 1 obtained by mixing  
3 4-chloro-1,8-naphthalic anhydride with monoamine  
4 polyether.

12. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 11 wherein said 4-  
3 chloro-1,8-naphthalic anhydride comprises about  
4 2% by weight of said 4-amino-1,8-naphthalimide  
5 compound and said monoamine comprises about 97%  
6 by weight of said 3 or 4-amino-1,8-naphthalimide  
7 compound.

13. The 3 or 4-amino-1,8-naphthalimide  
2 compound as recited in Claim 11 wherein said  
3 mixture is heated to approximately 150°C for 48  
4 to 72 hours.

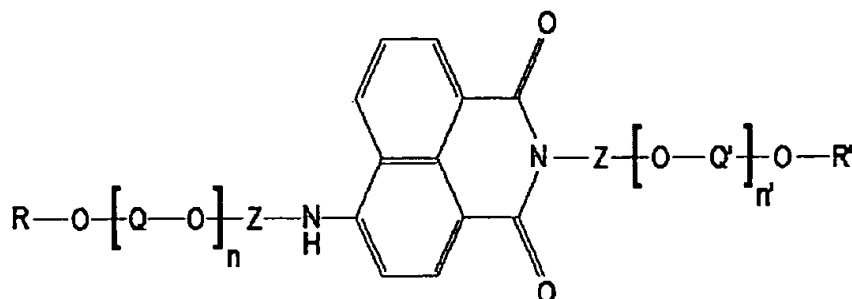


14. A method of detecting the fluorescent  
2 emission spectrum of a detection mixture,  
3 comprising the step of:  
4 mixing 3 or 4-amino-1,8-naphthalimide a with  
5 polyether substituent to form a mixture having  
6 polyether substituent groups at a 3 or 4-position  
7 and at an imide position, said polyether  
8 substituent groups linked to a hydrocarbon  
9 directly linked at said 3 or 4- position and said  
10 imide position, said polyether substituent groups  
11 rendering said 3 or 4-amino-1,8-naphthalimide  
12 compound sufficiently soluble in a fluid such  
13 that visualization of the fluid by color or  
14 fluorescence is allowed;  
15 mixing said mixture with said fluid to form  
16 said detection mixture;  
17 irradiating said detection mixture with a  
18 light sufficient to cause said detection mixture  
19 to emit a detectable excited wavelength emission  
20 spectrum; and  
21 detecting said emission spectrum of said  
22 detection mixture.

15. The method as recited in Claim 14  
2 wherein said step of mixing said mixture with  
3 said fluid includes the step of mixing said  
4 mixture with a polyalkylene glycol lubricant.

16. The method as recited in Claim 15  
2 wherein said polyalkylene glycol is  
3 polyethyleneglycol.

17. The method as recited in Claim 14  
 2 wherein said 3 or 4-amino-1,8-naphthalimide is a  
 3 4-amino-1,8-naphthalimide compound and has the  
 4 following general chemical structure:



5 where:

6 R or R' may be hydrogen, hydrocarbon, or  
 7 halogenated hydrocarbon and where R may or may  
 8 not equal R';

9 Q or Q' may be hydrocarbon or halogenated  
 10 hydrocarbon and where Q<sub>n</sub> may or may not be equal  
 11 to any other Q or Q'; and

12 Z or Z' may be hydrocarbon or halogenated  
 13 hydrocarbon and where Z may or may not equal Z'.

18. The method as recited in Claim 17  
 2 wherein n ranges from 0 to 200.

19. The method as recited in Claim 18  
 2 wherein n ranges from 9 to 10.

20. The method as recited in Claim 1  
 2 wherein said 3 or 4-amino-1,8-naphthalimide is  
 3 hydrophillic.

21. The method as recited in Claim 14  
2 wherein said hydrocarbon is a halogenated  
3 hydrocarbon.

22. The method as recited in Claim 14  
2 wherein said step of detecting includes the step  
3 of detecting said emission spectra in a range  
4 equal to or greater than 400 nm.

23. The method as recited in Claim 14  
2 wherein said substituents groups are polyether  
3 groups that have the following general formula:



5 where n may range from 5 to 15.

24. The method as recited in Claim 14  
2 wherein the step of mixing to obtain a mixture  
3 includes the step of mixing 4-chloro-1,8-  
4 naphthalic anhydride with monoamine polyether.

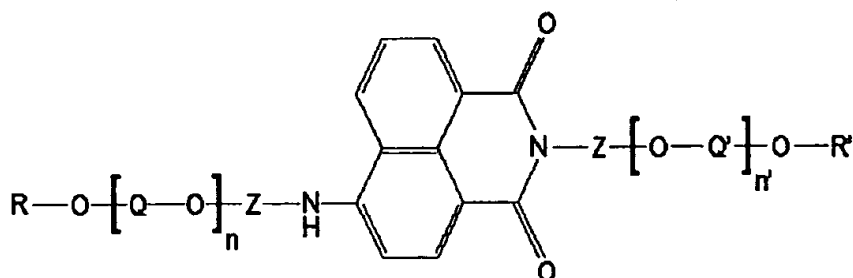
25. The method as recited in Claim 24  
2 wherein said step of mixing to obtain a mixture  
3 includes the step of mixing 4-chloro-1,8-  
4 naphthalic anhydride to comprise about 2% by  
5 weight of said mixture and said monoamine to  
6 comprise about 97% by weight of said mixture.

26. The method as recited in Claim 14  
2 further comprising the step of heating said  
3 mixture to approximately 150°C for 48 to 72  
4 hours.

27. An amino-1,8-naphthalimide compound  
2 being soluble in a fluid and having substituent  
3 groups at an imide position and a position distal  
4 to said imide position, said substituent groups  
5 linked to a hydrocarbon directly linked at said  
6 imide position and said distal position, said  
7 substituent groups having chemical properties  
8 that render said amino-1,8-naphthalimide compound  
9 sufficiently soluble in said fluid such that  
10 visualization of the fluid by color or  
11 fluorescence is allowed.

28. The amino-1,8-naphthalimide compound as  
2 recited in Claim 27 wherein said fluid is a  
3 polyalkylene glycol lubricant.

29. The amino-1,8-naphthalimide compound as recited in Claim 27 wherein said amino-1,8-naphthalimide is a 3 or 4-amino-1,8-naphthalimide compound that has the following general chemical structure:



6 where:

7 R or R' may be hydrogen, hydrocarbon, or  
8 halogenated hydrocarbon and where R may or may  
9 not equal R';

10 Q or Q' may be hydrocarbon or halogenated  
11 hydrocarbon and where Q<sub>n</sub> may or may not be equal  
12 to any other Q or Q'; and

13 Z or Z' may be hydrocarbon or halogenated  
14 hydrocarbon and where Z may or may not equal Z'.

30. The amino-1,8-naphthalimide compound as recited in Claim 27 wherein said amino-1,8-naphthalimide compound when combined with said fluid has a excitation spectra of equal to or greater than 400 nm.

31. The amino-1,8-naphthalimide compound as  
2 recited in Claim 27 wherein said substituent  
3 groups are polyether groups that have the  
4 following general formula:

5  $\text{CH}_3\text{--[O-CH}_2\text{-CH(CH}_3\text{)]}_n\text{--}$   
6 where n may range from 5 to 15.

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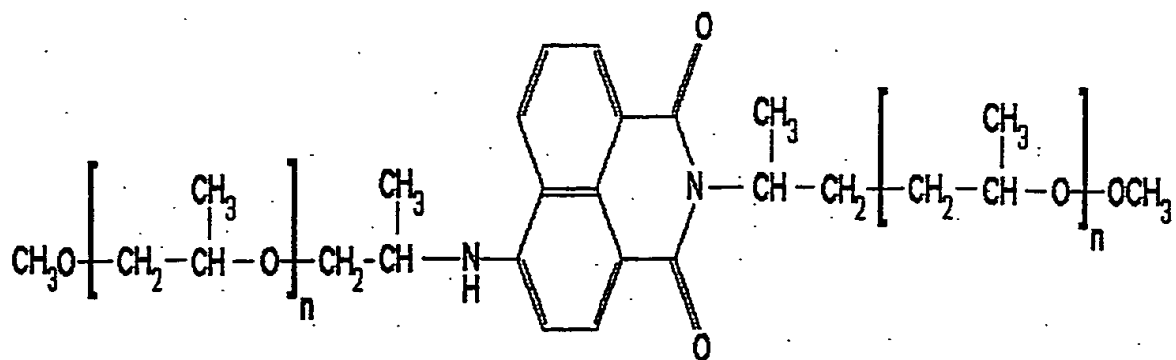


FIGURE 1

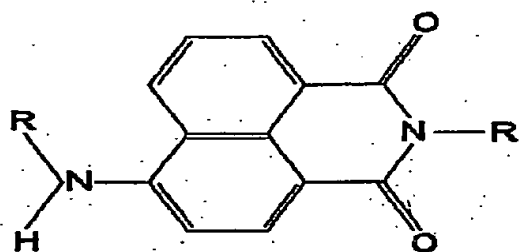


FIGURE 2

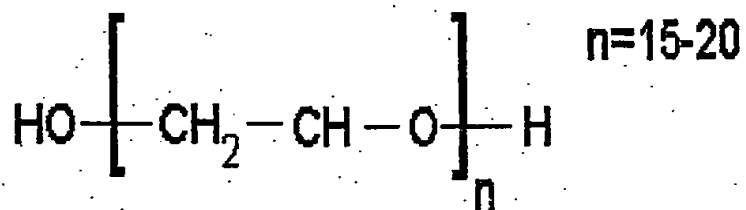


FIGURE 3

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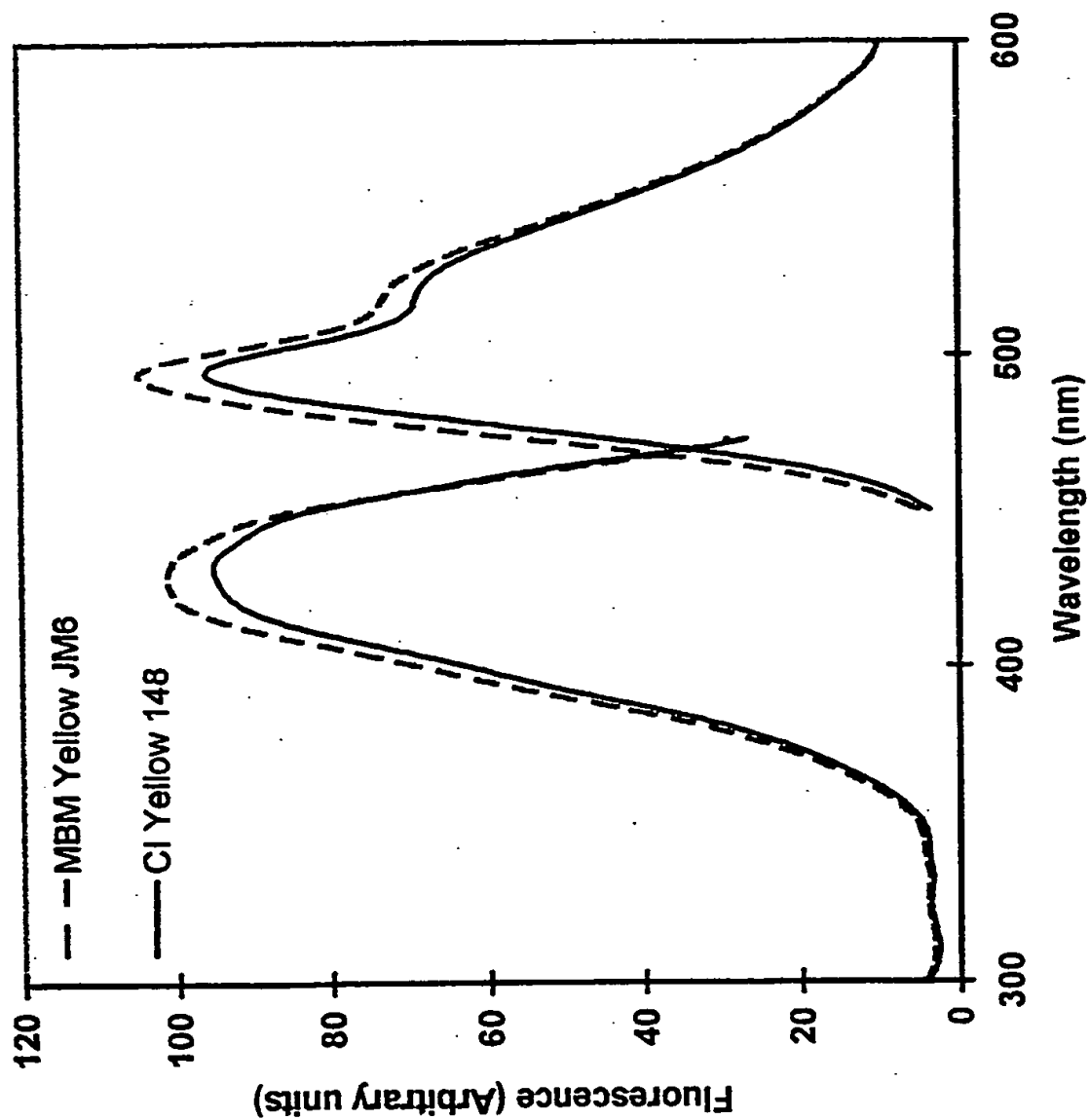


FIGURE 4



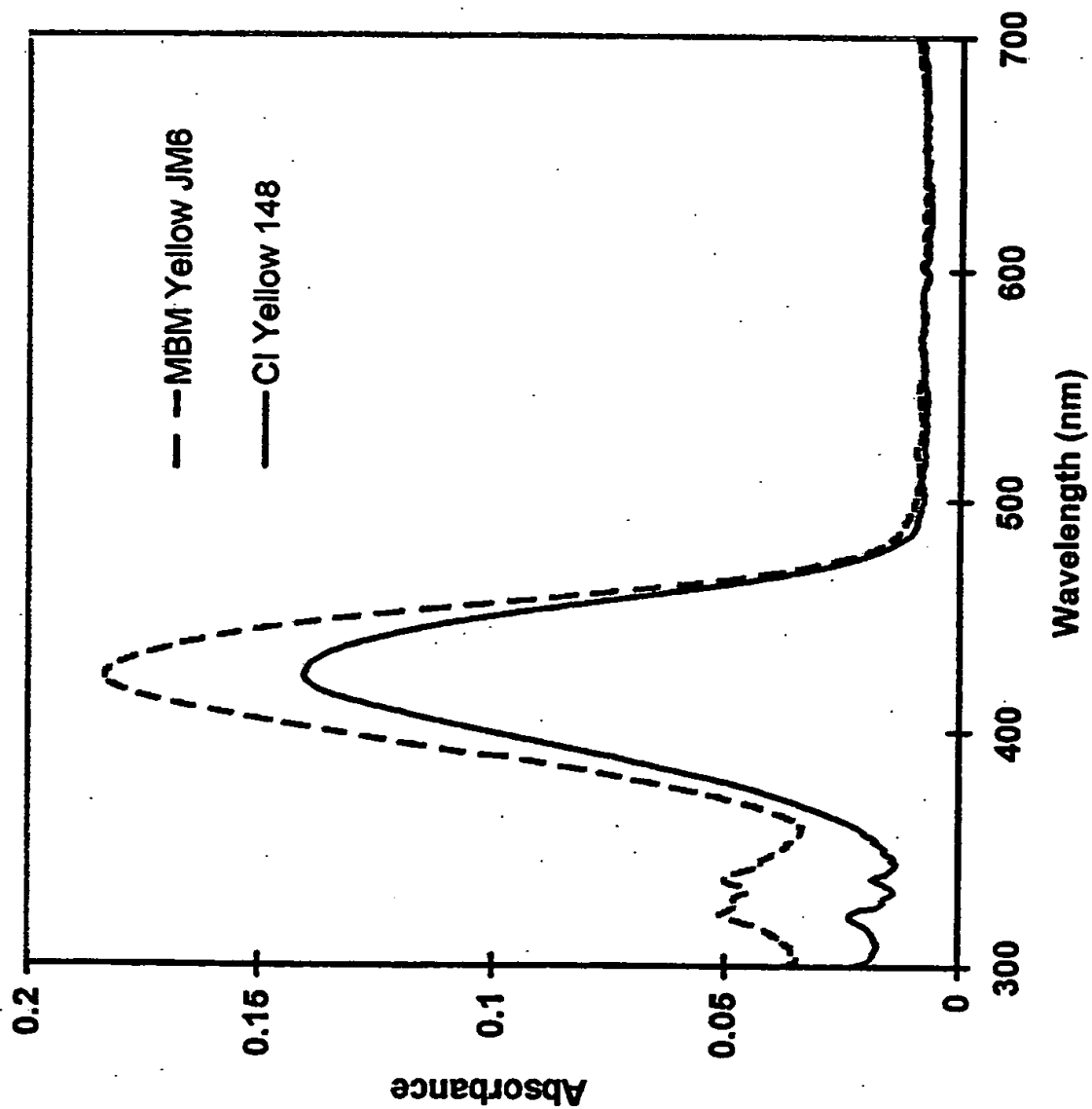


FIGURE 5

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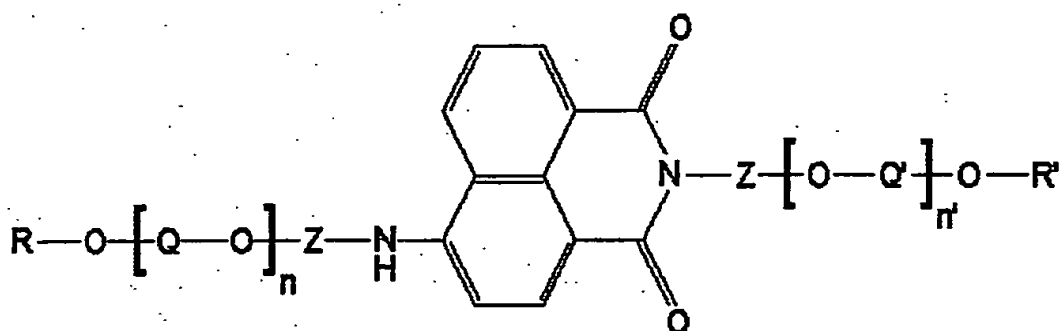


FIGURE 6

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/US 98/00325

**A. CLASSIFICATION OF SUBJECT MATTER**  
IPC 6 C09B69/00 G01M3/20

According to International Patent Classification(IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C09B G01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	EP 0 529 162 A (MILLIKEN RES CORP) 3 March 1993 see examples 71,71A	1,27
X	EP 0 437 105 A (MILLIKEN RES CORP) 17 July 1991 see examples 62,62A	1,27
A	US 5 235 045 A (LEWIS DAVID E ET AL) 10 August 1993 see claims 1-10	1-31
A	US 5 421 192 A (HENRY RICHARD G) 6 June 1995 see column 3, line 35 - line 64; claims	1-31

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search

24 April 1998

Date of mailing of the international search report

14/05/1998

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Ginoux, C

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

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